PATENT SPECIFICATION

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DRAWINGS ATTACHED

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(54) IMPROVEMENTS RELATING TO THE PRODUCTION OF CERAMIC SHEETS

(71) We, INTERNATIONAL BUSINESS MACHINES CORPORATION, a Corporation organized and existing under the laws of the State of New York in the United States of America, of Armonk, New York 10504, United States of America (assignees of WILFRED KARL NEUFFER) do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a method of forming a ceramic sheet.

Green ceramic sheets customarily are produced by processing particulate material slips by doctor blade casting or extrusion. After the green sheets are fired, they exhibit shrinkage anisotropy which is a function of the processing technique employed. Generally, shrinkage in the processing direction (the direction in which the material was moved) is less than in the transverse direction. Thus, it becomes necessary to make allowance for the anticipated shrinkage anisotropy in cases where close final dimensional tolerances are to be met. One example is in the multilayer ceramic circuit technology where high circuit density is achieved by stacking a plurality of interconnected circuit-bearing ceramic substrates. The establishment of electrical interconnections between the stacked substrates requires that close registration be maintained between the individual ceramic substrates during and after firing. Conventional practice requires that the processing directions of the individual ceramic sheets be remembered until the stacked assembly is made. If this were not done, the shrinkage anisotropy of the randomly aligned individual sheets would cause the loss of the vertical registration in the stacked assembly required for the electrical interconnections.

An object of the present invention is to provide an improved method of producing ceramic sheets.

According to the invention, a method of producing ceramic sheets comprises forming particulate material slip to produce a green ceramic sheet having a characteristic shrinkage anisotropy after firing, calendering said green ceramic sheet to induce in the sheet a shrinkage anisotropy after firing opposite in sense and substantially equal to said characteristic shrinkage anisotropy, and firing the calendered sheet.

The method of the present invention is based upon the discovery that a reduction in thickness of the green ceramic sheets by calendering induces a shrinkage anisotropy of the sheets after firing which is opposite in sense to the shrinkage anisotropy after firing resulting from the processing technique by which the green ceramic sheet is formed. In other words the shrinkage anisotropy induced by calendering is orthogonal to that induced by doctor blade techniques. The relationship between thickness reduction and shrinkage anisotropy can be experimentally determined by calendering a sample portion of a green ceramic sheet to produce a given thickness reduction. The calendered sample portion is then fired and a measure of the resulting shrinkage anisotropy is noted. A second sample portion is calendered in a similar manner except that a different thickness reduction is imparted to the second sample portion. A measure of the resulting shrinkage anisotropy after firing is noted for the second sample. In this manner, a desired number of data points are ascertained describing the relationship between thickness reduction and the measure of shrinkage anisotropy after firing. It has been found that the said relationship is substantially linear. Thus, the relationship defined by the experimentally derived data points may be extrapolated to predict the amount of required thickness reduction to achieve substantially zero shrinkage anisotropy after firing. The remaining green ceramic sheet represented by the sample portion is then calendered to produce the predicted thickness reduction thereby producing a sheet having no anisotropy after firing.

The invention will now be described, by way of example, with reference to the accompanying drawings, in which:—

Figure 1 is a simplified schematic of apparatus for doctor blading green ceramic sheet,

Figure 2 is a simplified schematic of apparatus for calendering processed green ceramic sheet, and

Figure 3 is a pair of typical plots describing the relationship between thickness reduction and percentage shrinkage anisotropy of fired green ceramic sheets.

Referring to Figure 1, particulate material slip 1 is cast by stationary doctor blade 2 onto flexible moving tape support 3. Support 3 may be, for example, a Mylar (Registered Trade Mark) polyester film. The flexible support 3 moves from supply reel 4 under idler wheel 5 and over support table 6 in the direction of arrow 7. Slip 1 in a representative case comprises a pulverized ceramic raw material such as alumina in an organic binder consisting of a solvent a wetting agent, a plasticizing agent, and a polymerizable resin, all of which are milled to form a homogeneous suspension. After slip 1 is cast, that is spread and levelled by doctor blade 2, into a thin sheet or film typically of 3 to 20 mils thickness on support 3, it is dried. The dried cast sheet is peeled from support 3 and allowed to stand until substantially all of the volatile constituents have evaporated from the sheet. The dried sheet, referred to as being in its green state, is now ready for further processing.

Green ceramic sheet not only can be produced by the disclosed doctor blading technique but also may be made by other conventional processes including the die and the roll extrusion of a highly viscous ceramic paste, the extrusion of an alkaline solution through a narrow slit, and the hot rolling of ceramic powder pre-mixed with thermal plastic binders. Irrespective of the method of fabrication, all green ceramic sheets have the common characteristic of shrinkage anisotropy after the sheet is fired. Generally, the shrinkage in the cast (processing) direction (designated X in Figure 1) is smaller than the shrinkage in the transverse direction (designated Y).

Shrinkage anisotropy is believed to be caused by the orientation of the ceramic particles during the casting process. Such orientation results from passing the ceramic slurry of slip through the narrow slit or gap characteristic of the various casting methods. Various devices and schemes have been used to reduce shrinkage anisotropy in the fired green ceramic sheets. According to one technique the shape of the doctor blade and the material used in its construction are designed and selected to reduce shrinkage anisotropy. Other techniques include the application of high frequency electrical energy to the area of casting to disturb the particle orientation during the casting process. These methods, however, are not applicable to those casting techniques which require the use of high viscosity mixes for the slip.

In accordance with the method of the present invention, the dried green ceramic sheets are calendered to substantially compensate for the shrinkage anisotropy after firing resulting from the casting process. Substantially zero shrinkage anisotropy after firing may be achieved by calendering at any time after the green ceramic sheets have been produced and dried. Successful results have been achieved, for example, where the dried ceramic sheets were several weeks old. Inasmuch as the green ceramic sheets are calendered after they have been produced and dried, the method is applicable for the substantial elimination of shrinkage anisotropy of fired green ceramic sheets irrespective of the manner in which they were made.

Referring to Figure 2, dried green ceramic sheet 8 is driven in the direction of arrow 9 through calendering rollers 10 and 11. Depending upon the diameter of the calendering rollers, the roller pressure, the process temperature, and the composition of the sheet 8, a shrinkage anisotropy after firing is induced which is opposite that attributable to the casting process. That is, the shrinkage of the fired calendered sheet in the processing direction designated X is greater than the shrinkage in the transverse direction designated Y.

Calendering elongates as well as densifies the green ceramic sheets. Both effects combine to induce the desired compensatory shrinkage anisotropy. It has been found, however, that the shrinkage anisotropy induced by calendering is more quickly achieved by elongating the green ceramic sheet rather than densifying it. Elongation cannot be achieved without densification and vice versa, but smaller diameter calendering rolls maximise the elongation and minimize the densification whereas larger diameter calendering rools tend to densify rather than elongate.

Figure 3 illustrates typical plots showing the relationship between percentage shrinkage anisotropy, defined as the difference between the percentage shrinkages after firing in the X and Y direction referred to previously, and percentage sheet thickness reduction for small diameter rolls (curve 12) and larger diameter rolls (curve 13) respectively. It should be observed that although both relationships are substantially linear, the slope of curve 12 is significantly greater than that of curve 13. For example, zero shrinkage anisotropy after firing is achieved with a 6% thickness reduction using the small diameter rolls whereas a 13% thickness reduction is required for the same result using the larger diameter calendering rolls.

It may not be necessary to determine experimentally the complete function such as plotted in Figure 3 in order to ascertain the percentage

thickness reduction required to achieve substantially zero shrinkage anisotropy. It is sufficient to determine experimentally any representative portion of the function and to extrapolate analytically the function to the desired shrinkage coordinate value and the corresponding thickness reduction coordinate value. The remaining green ceramic sheet represented by the sample portions used in the experiments is then calendered to produce the analytically extrapolated thickness reduction value.

WHAT WE CLAIM IS:—

1. A method for producing ceramic sheet comprising forming particulate material slip to produce a green ceramic sheet having a characteristic shrinkage anisotropy after firing, calendering said green ceramic sheet to induce in the sheet a shrinkage anisotropy after firing opposite in sense and substantially equal to said characteristic shrinkage anisotropy, and firing the calendered sheet.

2. A method as claimed in claim 1, wherein said particular material slip is formed by doctor blade casting.

3. A method as claimed in claim 1 or claim 2, in which the green sheet is calendered by an amount determined by calendering a plurality of sample portions of said sheet to produce a respective thickness reduction in each sample portion, firing each calendered sample portion, measuring the percentage shrinkage anisotropy (as hereinbefore defined) in each fired calendered sample portion to determine the relationship between thickness reduction and percentage shrinkage anisotropy after firing the characteristic of said sample portions, and extrapolating said relationship, if necessary, to determine the thickness reduction necessary to achieve a substantially zero shrinkage anisotropy.

4. A method of producing ceramic sheet, substantially as herein described.

5. Ceramic sheet produced by a method claimed in any preceding claim.

JOHN BLAKE,
Chartered Patent Agent,
Agent for the Applicants.

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1277695 COMPLETE SPECIFICATION

1 SHEET *This drawing is a reproduction of the Original on a reduced scale*

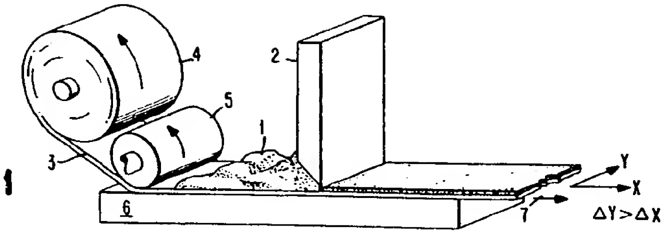

FIG. 1

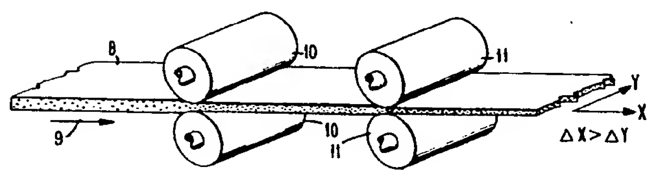

FIG. 2

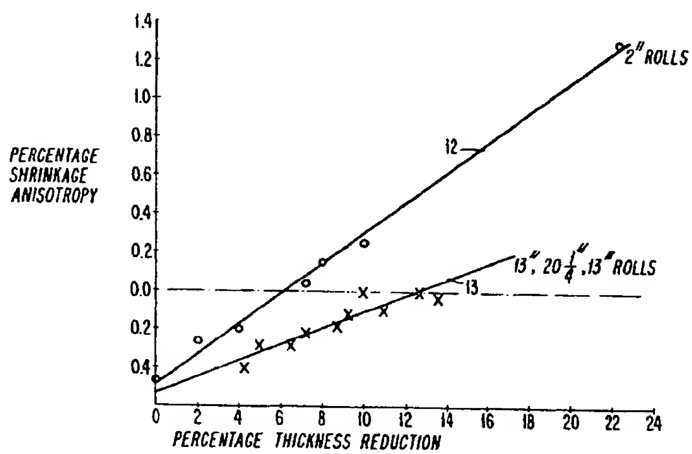

FIG. 3